Surface Free Energies and Surface Entropies of Pure Liquid Metals

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The surface free energy or surface tension, of a liquid-gas interface may be described theoretically using the two statistical mechanical approaches employed for the equilibrium properties of liquids: the radial distribution function and the free-volume method. Calculations using the former method have been carried out by Fowler, Kirkwood and Buff; the latter method has been used by Lennard-Jones and Corner, Prigogine and Saraga. Johnson, Hutchinson and March and others have calculated the surface free energy values of low melting-point metals using experimental data available for pair distribution function and pair potential through the use of Fowler's expression. Owing to the poor agreement between calculation and experiment, a close examination on the accuracy of pair distribution functions on the liquid-gas interface and the validity of Fowler's approximation is needed. However, the free-volume method has not, as yet, been applied to liquid metals, due to the difficulty of describing the liquid structure model and its potential model. In the present paper, in order to evaluate the surface free energy of liquid metals, a new thermodynamic approximate-expression for surface free energy was proposed on the basis of the surface free energy from the free-volume method, by using additive rules for the evaluation of interaction energy and free volume on surface atoms. By regarding liquid metals as having tetrahedral and octahedral structure, triangular-square-pentagonal lattices were proposed for a surface structural model. A comparison between calculated values for surface free energy and surface entropy and their previous experimental data in 7 liquid metals indicated the validity of the thermodynamic approximate-expression for the surface free energy and the surface structural model, over a wide temperature-range including an undercooling region.